

1470/c

PATENT SPECIFICATION

DRAWINGS ATTACHED

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824,789



Date of filing Complete Specification Jan. 15, 1958.

Application Date Jan. 15, 1957.

No. 1458/57.

Complete Specification Published Dec. 2, 1959.

Index at acceptance:—Class 40(3), A5(M3:R8).

International Classification:—G08c.

COMPLETE SPECIFICATION

Improvements in or relating to methods of and means for Radioactivation Analysis

We, UNITED KINGDOM ATOMIC ENERGY AUTHORITY, London, a British Authority, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to methods of and means for radioactivation analysis.

The concentration of trace elements in a base material can often be measured by radioactivation analysis, in which the whole sample is irradiated with neutrons and the resulting radiations then measured. By using a scintillation counter and pulse amplitude analyser (kicksorter) to measure gamma rays of a particular energy characteristic of each trace element, the radiations can sometimes be measured selectively without the need for chemical separation.

Accurate analysis becomes difficult when the range of pulse amplitudes selected by the kicksorter includes those of pulses arising from radiations of the base material. Such pulses may arise either (a) from the continuous spectrum of recoil electrons corresponding to the Compton Scattering of higher energy gamma-rays within the phosphor, or (b) from gamma rays emitted by other constituents of the base material, whose energies are too close to the required energy to be resolved.

It has already been shown (Peirson, Brit. J. Appl. Phys. vol. 6, p. 444, 1955) that the spectrum of recoil electrons can be eliminated by observing a source simultaneously with two scintillation counters, using NaI(Tl) and anthracene respectively as phosphors. By suitable matching, the output of the anthracene counter (which records the recoil spectrum only) can be subtracted from that of the NaI(Tl) counter, to leave only the photoelectric responses of the latter.

In many practical problems, the element to

be analysed is an additive, and samples of the base material without the additive are available. Assay is simple when the additive can be made radioactive before addition, but this is not always practicable. It is an object of the present invention to provide a method of and means for determining the concentration of a trace element in a base material when samples of the base material without the trace element, or samples of the constituent elements of the base material, are available.

According to the present invention a method of determining the concentration of a specified element in the presence of one or more other elements comprises deriving simultaneously electrical outputs corresponding to the gamma-ray spectra of a first irradiated sample of the mixture of elements including the specified element, and of a second irradiated sample excluding said specified element but including at least one of said other elements, the difference between said outputs being recorded whereby the resultant spectrum effectively isolates the photoelectric peak or peaks of the specified element.

The second sample may consist of a further irradiated quantity of an original mixture to which the specified element was added. Alternatively the second sample may be composed of pure irradiated samples of the constituent elements of the mixture, successively added until the photoelectric peak or peaks of the specified element are effectively isolated.

The concentration of the specified element may be estimated directly from the height of its photoelectric peaks, or may be determined by adding a measured amount of the irradiated trace element to the second sample until its peak or peaks are suppressed.

Also according to the present invention, means for determining the concentration of a specified element in the presence of one or more other elements comprises two similar

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scintillation counters having phosphors with matched energy resolution and responsive to photoelectric absorption, means whereby each of two sources may be located at a variable distance from each of said counters, gamma-ray shielding means whereby radiation from each said source is received by only one of said counters, and means whereby the gamma-ray spectra of the two sources are derived simultaneously, subtracted and recorded.

The phosphors may be thallium-activated sodium iodide crystals.

To enable the nature of the invention to be more readily understood, attention is directed towards the drawings accompanying the provisional specification wherein:—

Fig. 1 is a block schematic diagram of an embodiment of the invention; and

Figs. 2 and 3 show spectra plotted there-with.

In Fig. 1 similar photomultiplier tubes 1 and 2 are shown with their photo-cathodes exposed to similar matched thallium-activated sodium iodide crystals 3 and 4 respectively. Crystals 3 and 4 are exposed to sources 5 and 6 respectively whose distance from their respective crystals can be varied. A gamma-ray shield 7 prevents radiation from source 5 reaching crystal 4 and radiation from source 6 reaching crystal 3. The photomultipliers are rendered inoperative in turn by a switch 8 which earths their first dynodes, so that an amplifier 9 receives the output of each in turn. The switch 8 is operated by a multi-vibrator (not shown) which generates a 0.5 sec duration square wave. The amplifier output is fed to a single-channel pulse amplitude analyser 13 whose discriminator bias is varied by a potentiometer ganged to the driving mechanism of a recorder 10. The output of the kicksorter is connected through a switch 11, operated in synchronism with the switch 8, to a double ratemeter 12. The output of this ratemeter is proportional to the difference between the pulse rates at the two inputs. The above circuit arrangement is the same as that described by Peirson in the paper already referred to, except that both of the present scintillation counters detect photoelectric scintillators.

In operation one of the sources (source 5 for example) consists of an irradiated sample of a mixture of elements including the element to be determined. In one method of determination the other source (source 6 in this example) consists of an irradiated sample of the same mixture excluding the element, the latter having been an additive to the mixture. The HT voltages on the two counters are adjusted to produce equal pulse amplitudes for gamma-rays of equal energies and the source distances are adjusted to match counting rates. Under these conditions the difference between the gamma-ray spectra of the two samples plotted by the equipment, which is displayed by the

recorder, consists of the spectrum of the added element only.

An example of such a measurement is shown in Fig. 2. The object was to determine the amount of selenium decolouriser which had been added to some glass. Samples of the glass with and without the selenium additive were irradiated in a nuclear reactor and their spectra plotted. The counter HT's and source distances were adjusted to match pulse amplitudes and counting rates respectively at an energy above those of the Se^{75} gamma-rays. Curve (c) shows the resultant spectrum and curve (d) shows by comparison the spectrum obtained from a pure Se^{75} source. From the latter the selenium content of the glass (17 p.p.m.) could be deduced. Curves (a) and (b) show the complete spectra of the glass containing the added element and of the glass without the added element respectively, which are subtracted to produce the spectrum (c). The difference in selenium content as deduced from the two individual peaks (at 0.138 and 0.269 MeV) is 5%, indicating that the error in balancing is small, though it does introduce an increased statistical fluctuation.

A null method of determining the selenium content is to add a measured quantity of Se^{75} to the balancing source of glass without additive until the Se^{75} peaks disappear.

Fig. 3 shows how an unwanted spectrum in a mixture can be suppressed using a separate irradiated sample of the constituent responsible for the unwanted spectrum. Curve (a) is the record obtained with a Hg^{203} source exposed to one of the counters, showing the 0.28 MeV photoelectric peak. Without moving the Hg^{203} source, a Se^{75} source was then added to it. This combined $\text{Hg}^{203}/\text{Se}^{75}$ source simulated an irradiated mixture of mercury and selenium in which it was desired to determine the quantity of mercury. Curve (b) shows the resulting composite spectrum, in which the 0.27 MeV peak of Se^{75} was superimposed on, and completely masked, the Hg^{203} peak, so making it impossible to determine the Hg^{203} content. Another Se^{75} source was then applied to the second counter and balance obtained by adjusting the source distance until the isolated 0.45 MeV Se^{75} peak disappeared. Curve (c) shows the Hg^{203} peak as it appeared after subtraction. Agreement between the areas under the Hg^{203} peaks in curves (a) and (c) is within 7%. The method can be extended to suppress several unwanted spectra by introducing successive irradiated samples of the appropriate constituents at the balancing counter, provided isolated peaks are available for balancing out.

WHAT WE CLAIM IS:—

1. A method of determining the concentration of a specified element in the presence of one or more other elements comprising deriving simultaneously electrical outputs corresponding to the gamma-ray spectra of a first

irradiated sample of the one or more other elements and the specified element, and of a second irradiated sample excluding said specified element but including at least one of said other elements, the difference between said outputs being recorded whereby the resultant spectrum effectively isolates the photoelectric peak or peaks of the specified element.

2. A method as claimed in Claim 1 wherein said one or more other elements constitute a material to which the specified element has been added, and the second sample is an irradiated sample of said material excluding said specified element.

3. A method as claimed in Claim 1 wherein the second sample comprises separate irradiated samples of at least one of said one or more other elements.

4. A method as claimed in any preceding claim comprising adding measured amounts of the irradiated specified element to the second sample until the peak or peaks associated with the specified element are suppressed.

5. Means for determining the concentration

of a specified element in the presence of one or more other elements comprising two similar scintillation counters having phosphors with matched energy resolution and responsive to photoelectric absorption, means whereby each of two sources may be located at a variable distance from each of said counters, and gamma-ray shielding means whereby radiation from each said source is received by only one of said counters, whereby the gamma-ray spectra of the two sources can be derived simultaneously, subtracted and recorded.

6. Means as claimed in Claim 5 wherein the phosphors are thallium-activated sodium iodide crystals.

7. Means for determining the concentration of a specified element in the presence of one or more elements substantially as hereinbefore described with reference to the drawings accompanying the provisional specification.

S. C. BARTLETT,
Chartered Patent Agent,
Agent for Applicants.

PROVISIONAL SPECIFICATION

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The concentration of trace elements in a base material can often be measured by radioactivation analysis, in which the whole sample is irradiated with neutrons and the resulting radiations then measured. By using a scintillation counter and pulse amplitude analyser (kicksorter) to measure gamma rays of a particular energy characteristic of each trace element, the radiations can sometimes be measured selectively without the need for chemical separation.

Accurate analysis becomes difficult when the range of pulse amplitudes selected by the kick-sorter includes those of pulses arising from radiations of the base material. Such pulses may arise either (a) from the continuous spectrum of recoil electrons corresponding to the Compton Scattering of higher energy gamma-rays within the phosphor, or (b) from gamma rays emitted by other constituents of the base material, whose energies are too close to the required energy to be resolved.

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counter (which records the recoil spectrum only, can be subtracted from that of the NaI(Tl) counter, to leave only the photoelectric responses of the latter.

In many practical problems, the element to be analysed is an additive, and samples of the base material without the additive are available. Assay is simple when the additive can be made radioactive before addition, but this is not always practicable. It is an object of the present invention to provide a method of and means for determining the concentration of a trace element in a base material when samples of the base material without the trace element, or samples of the constituent elements of the base material, are available.

According to the present invention a method of determining the concentration of a specified element in the presence of one or more other elements comprises deriving simultaneously electrical outputs corresponding to the gamma-ray spectra of a first irradiated sample of the mixture of elements including the specified element, and of a second irradiated sample excluding said specified element but including at least one of said other elements, the difference between said outputs being recorded whereby the resultant spectrum effectively isolates the photoelectric peak or peaks of the specified element.

The second sample may consist of a further irradiated quantity of an original mixture to which the specified element was added. Alternatively the second sample may be composed of pure irradiated samples of the constituent

elements of the mixture, successively added until the photoelectric peak or peaks of the specified element are effectively isolated.

5 The concentration of the specified element may be estimated directly from the height of its photoelectric peaks, or may be determined by adding a measured amount of the irradiated trace element to the second sample until its peak or peaks are suppressed.

10 Also according to the present invention, means for determining the concentration of a specified element in the presence of one or more other elements comprises two similar scintillation counters having phosphors with matched energy resolution and responsive to photoelectric absorption, means whereby each of two sources may be located at a variable distance from each of said counters, gamma-ray shielding means whereby radiation from each source is received by only one of said counters, and means whereby the gamma-ray spectra of the two sources are derived simultaneously, subtracted and recorded.

25 The phosphors may be thallium-activated sodium iodide crystals.

To enable the nature of the invention to be more readily understood, attention is directed towards the accompanying drawings wherein:—

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for example) consists of an irradiated sample of a mixture of elements including the element to be determined. In one method of determination the other source (source 6 in this example) consists of an irradiated sample of the same mixture excluding the element, the latter having been an additive to the mixture. The HT voltages on the two counters are adjusted to produce equal pulse amplitudes for gamma-rays of equal energies and the source distances are adjusted to match counting rates. Under these conditions the difference between the gamma-ray spectra of the two samples plotted by the equipment, which is displayed by the recorder, consists of the spectrum of the added element only.

80 An example of such a measurement is shown in Fig. 2. The object was to determine the amount of selenium decolouriser which had been added to some glass. Samples of the glass with and without the selenium additive were irradiated in a nuclear reactor and their spectra plotted. The counter HT's and source distances were adjusted to match pulse amplitudes and counting rates respectively at an energy above those of the Se^{75} gamma-rays. Curve (c) shows the resultant spectrum and curve (d) shows by comparison the spectrum obtained from a pure Se^{75} source. From the latter the selenium content of the glass (17 p.p.m.) could be deduced. Curves (a) and (b) show the complete spectra of the glass containing the added element and of the glass without the added element respectively, which are subtracted to produce the spectrum (c). The difference in selenium content as deduced from the two individual peaks (at 0.138 and 0.269 MeV) is 5%, indicating that the error in balancing is small, though it does introduce an increased statistical fluctuation.

A null method of determining the selenium content is to add a measured quantity of Se^{75} to the balancing source of glass without additive until the Se^{75} peaks disappear.

Fig. 3 illustrates the method by which a composite source is used to suppress unwanted spectra. Curve (a) is the record obtained with a Hg^{203} source exposed to one of the counters, showing the 0.28 MeV photoelectric peak. Without moving the Hg^{203} source, a Se^{75} source was then added to it. Curve (b) shows the resulting composite spectrum, in which the 0.27 MeV peak of the Se^{75} is superimposed, completely obliterating the Hg^{203} peak. Another Se^{75} source was then applied to the second counter and balance obtained by adjusting the source distance until the 0.45 MeV Se^{75} peak disappeared. Curve (c) shows the Hg^{203} peak as it appeared after subtraction. Agreement between areas under the Hg^{203} peaks is within 7%. The method may be extended to suppress several unwanted spectra

by introducing successive pure samples at the
balancing counter.

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Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press.—1959.
**Published at The Patent Office, 25, Southampton Buildings, London, W.C.2, from which
copies may be obtained.**

